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TRANSFER OF RADIONUCLIDES OF THE URANIUM AND THORIUM DECAY CHAINS IN AQUATIC AND TERRESTRIAL ENVIRONMENTS

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ABSTRACT

This report examines the transfer of radionuclides from the uranium and thorium decay chains (U-238, Ra-226, Th-232, Th-230, Po-210 and Pb-210) through the aquatic and terrestrial environment. This transfer is characterized by a transfer coefficient; environmental and experimental factors which cause this coefficient to vary are presented and discussed in this report. Furthermore, based on a literature survey, the report indicates the range of coefficients found for the aquatic sector (that is, sediment and freshwater and marine organisms) and for the terrestrial sector (that is, plants and domestic and wild animals). Afterwards, generalisations are formulated on the transfer of the different radionuclides through the multiple environmental compartments.

RÉSUMÉ

Ce rapport traite du transfert des radionucléides des chaînes de désintégration de l'uranium et du thorium (U 238, Ra 226, Th 232, Th 230, Po 210 et Pb 210) dans les milieux aquatique et terrestre. Ce transfert est caractérisé par le coefficient de transfert; les facteurs environnementaux et expérimentaux qui font varier ce coefficient sont discutés dans ce rapport. De plus, basé sur une revue bibliographique, le rapport indique l'éventail des coefficients trouvés et pour le milieu aquatique (soit le sédiment ainsi que les organismes d'eaux douces et marines) et pour le milieu terrestre (soit les plantes ainsi que les animaux domestiques et sauvages). Par la suite, des généralisations sont émises sur le transfert des différents radionucléides à travers les multiples composantes environnementales.

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INTRODUCTION

In Canada, the Atomic Energy Control Board is responsible for regulating the dose to members of the public from radioactive releases of nuclear facilities. Compliance with the dose limit is assessed by calculating the total dose received from different pathways such as inhalation and ingestion of radionuclides as well as from direct exposure. Generally, the transfer of radionuclides in the environment is estimated by means of appropriate mathematical models. These models use transfer coefficients[1]; ideally, site-specific information should be used in these models, however when this is not feasible, the coefficients can be obtained from the literature.

Several publications are available that can be used as sources of estimated coefficients. The International Atomic Energy Agency (IAEA, 1982), the United States Nuclear Regulatory Commission (USNRC, 1976), and the National Research Council of Canada (NRCC, 1983), are three examples of such sources. In addition, a Canadian publication prepared by the Canadian Standards Association (CSA), entitled Guidelines for Calculating Derived Release Limits for Radioactive Material in Gaseous and Liquid Effluent for Normal Operation of Nuclear Facilities (N288.1) will soon be available.

In the above publications, some of the proposed coefficients are very uncertain since they are derived from a very small number of studies, especially for those radionuclides in the uranium and thorium decay chains

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1. The transfer coefficient is also known as the concentration coefficient, the concentration factor, coefficient of bioaccumulation, or biological accumulation ratio (BAR) (Sheppard, 1980). This coefficient is the ratio between the concentration of radionuclides in the biological sample, and the concentration of radionuclides in the environmental medium. The definition of this coefficient is often modified for terrestrial domestic animals. It is then defined as the result of dividing the initial coefficient by the rate of ingestion of feed which is expressed in days per liter or days per kilogram.

which are released to the environment at the front end of the nuclear fuel cycle, i.e., uranium mines, mills, refineries and fuel fabrication plants. The purpose of this report is therefore to independently review the literature (emphasizing Canadian references) in order that AECB staff may evaluate the coefficients used by licensees in their environmental pathways analyses. As a corollary, licensees could use this report as a guide for estimating transfer coefficients when site-specific data are not readily available.

Chapter I

SOURCES OF VARIATION IN THE COEFFICIENTS

1.1 Biotic and Abiotic Factors

The review of the literature revealed a very wide variation of coefficients which depended on spatial and temporal conditions. Coefficients for terrestrial environments are more constant than those measured in aquatic environments, where there are very large spatial, and temporal fluctuations, especially in fresh waters. However, at this point, these findings cannot be confirmed, given the scarcity, and even the contradictory nature of the available data.

Table I shows the environmental (biotic and abiotic) factors that affect the values of these coefficients. They have been listed in two columns, according to whether they pertain to terrestrial or aquatic environments. The relative importance of these factors is little known, and it varies with the type of organism, the radionuclide under consideration, and the intensity of that factor in a particular environment. Before discussing these factors, it should be noted that a state of equilibrium is assumed between the concentrations of radionuclides in the medium and in the biological organism because of conditions resulting from chronic exposure. In the case of acute exposure, the coefficients are generally lower, since the organisms have not had time to adapt to the exposure (Krumholz *et al.*, 1957).

(a) Terrestrial Environment

There are many biotic and abiotic factors that affect the accumulation of radionuclides by terrestrial vegetation. The accumulation potential varies according to the species, and is affected by various biotic factors such as metabolism (Osborn, 1965); the root system (Osborn, 1965; Grzybowska, 1974); and the growth stage (Cannot, *in* Grzybowska, 1974). The higher the rate of growth of the plant, the larger the accumulation of radionuclides (Schreckhise and Cline, 1980; Kunacheva, 1944). The distribution of radionuclides in the plant also varies. For example, uranium is more concentrated in young tissue (e.g. seeds, the tips of leaves), while thorium is more concentrated in old

Table I The Main Biotic and Abiotic Factors that Affect the Accumulation of Radionuclides

Terrestrial Environment		Aquatic Environment	
Plants		**Sediment**	
Biotic factors	. type of species . root system . metabolism . stage and rate of growth . part of the plant . density of vegetation . ash content		. amount of oxygen . amount of dissolved organic carbon . alkalinity . temperature . flow rate
Abiotic factors	. solubility of radionuclide . soil pH . organic matter . physico-chemical form of the radionuclide . light . season . texture of the soil . concentration of the radionuclide in the soil . presence of analogous elements in the environment (antagonism, synergism, substitution)	**Plants**	
		Biotic factors	. type of species . biomass . accumulation pathways
		Abiotic factors	. presence of analogous elements in the environment (antagonism, synergism, substitution) . salinity . percentage of dissolved solids . concentration of radionuclide in water . pH of water . composition physico-chemical characteristics of the water . light . particles in suspension
Animals		**Animals**	
Biotic factors	. type of species . metabolism . diet . lactation stage . homeothermia (birds and mammals)	Biotic factors	. organ specificity . growth . osmotic and ionic regulation . nutritional mode . poikilothermia . accumulation pathways
Abiotic factors	. season . ingestion of soil	Abiotic factors	. see "aquatic plants"

tissue (e.g. wood) (Osborn, 1965). The density of vegetation is another important factor that affects the transfer of radionuclides, because the total amount of radionuclides is diluted by a larger biomass (Garten, 1972). Finally, Federov and Romanov (1969) have found that the accumulation of uranium correlates closely with the ash content of plants; however, the mechanism behind this correlation has not been studied in depth.

The effects of environmental factors have been largely studied in terms of the accumulation of radium by plants. The physico-chemical form of the radionuclide determines its solubility (Burkart, 1983), and its mobility (McDowell-Boyer et al., 1979) in the soil. It is also known that the lower the pH of the soil (acid soil), the more there is accumulation of radium by the vegetation (Kunasheva, 1944). Rayno (1982) has pointed out that the accumulation of radium may increase by up to one order of magnitude in soils that have a large concentration of organic matter. The texture of the soil is another important (if not the most important) factor affecting the accumulation potential. In general, the smaller the soil particles (clay) the larger the retention of radionuclides by the soil. For example, Ashkinazi (1982) found a coefficient of 0,02 for carrots growing in clay soil, and of 0,3 for potatoes growing in sandy soil. It is not mentioned to what extent these differences are due to the retention of radium in the soil, rather than to differences between species. The concentration of radionuclides in the soil also affects the amount of radionuclides absorbed by the plant. Sheppard (1980) points out that large concentrations of uranium in the soil facilitate a larger accumulation of uranium in the plant, except in boggy locations where there is less accumulation of uranium.

Furthermore, the presence of analogous elements in the environment can affect the accumulation of radionuclides. This phenomenon has been studied mainly in terms of the relationship that exists between calcium and radium. Even though the chemical activity of radium is different from that of calcium, their interaction with biological systems is similar, in that they compete for the same absorption and absorption sites. Burkart (1983) found a negative correlation between the presence of calcium in the soil and the accumulation of radium by the plants. Burkart (1983) suggests that a diet low in calcium

may prevent the accumulation of radium in humans. This relationship between radium and calcium has not been confirmed by all other authors; Grzybowska (1974), among others, did not find any link between these two elements. Thus, the competition between radium and calcium still remains to be clearly established.

Given that most terrestrial animals consumed by man are homeothermic, the accumulation of radionuclides in terrestrial animals is much less affected by the environment. Biotic factors, such as diet and the growth stage, have a larger effect on accumulation potential than abiotic factors, such as seasonal changes. For instance, the ingestion of soil by animals during grazing which is species specific can affect the transfer coefficients (grazing cattle consume up to 6% soil while lambs consume up to 10% (McDowell-Boyer et al., 1979)).

(b) Aquatic Environment

Sediments contain significant amounts of radionuclides, but there are still many unresolved questions regarding its capacity to accumulate radionuclides. A few studies have produced interesting results. Wahlgren and Orlandi (1981), in their study of some North American lakes, observed that the accumulation of thorium by sediment is larger in acidic systems containing large quantities of dissolved organic carbon, whereas the adsorption of uranium is lower in alkaline water, because of the competition between calcium and uranium for adsorption sites. A study conducted by Saskmont Engineering Ltd. (1983) revealed that a temperature increase facilitates the adsorption of radium, and that a strong current prevents the adsorption of radium by the sediment. Hesslein and Slavicek (1984) found that, in systems that are not heavily oxygenated, the components of radium tend to desorb in the water.

In aquatic systems, the accumulation of radionuclides by organisms is more affected by abiotic than by biotic factors. Bioaccumulation by aquatic plants is affected by abiotic factors, such as salinity, temperature, percentage of dissolved solids (Krumholz et al., 1957), the concentration of radionuclides in the water, the pH of the water (Ruggles and Rowley, 1978), alkalinity (Ophel, 1978), and light (Polikarpov, 1964).

With the exception of mammals, aquatic animals are poikilothermic, and their internal systems are affected by fluctuations in the environment. Nevertheless, certain biotic factors can have a direct effect on their capacity to accumulate radionuclides. Among these factors, there are growth, osmotic and ionic regulation, organ specificity, and diet (Krumholz et al., 1957). In fact, Kovalsky et al., (1967) have illustrated the latter by pointing out that predator fish, such as trout, accumulate radionuclides less than herbivorous fish, such as carp. Radionuclides are mainly accumulated by diffusion, either through the respiratory system (gills, filters), or through the ingestion of food or sediment. The relative importance of these channels is a function of the species under consideration, and remains to be clarified. At present, the calculation of transfer coefficients assumes that the uptake is larger by the respiratory system.)

With the exception of light, the abiotic factors affecting the accumulation potential of aquatic animals are the same as those that affect aquatic plants. In addition to these factors, the uptake of radionuclides by aquatic animals may be influenced by the turbidity of the water. It is known that the presence of particles in suspension slows down the growth of filter feeders (Létourneau, 1982; Fretter and Graham, 1962); it is therefore expected that there is less accumulation of radionuclides in turbid environments, however, this hypothesis remains to be proven.

Ophel (1978) points out that freshwater fish accumulate 10 times more radium and uranium in waters that are not very alkaline (> 150 mg Ca) than in environments that are strongly alkaline. In alkaline environments, a high pH increases the presence of the carbonate ion. This facilitates the formation of soluble uranyl carbonate, which is not accumulated by fish (Bondietti et al., 1979). The effect of calcium on the accumulation of radium has been observed in mollusks. In Australia, Jeffree and Davy (1983) found a negative correlation between the concentration of calcium in the environment and the accumulation of radium; the more calcium there is in the aquatic environment, the less is the accumulation of radium in the flesh of mollusk.

The concentration of radionuclides in the environment affects the rate of accumulation by aquatic animals (Polikarpov, 1964). This has been illustrated

by Martin et al. (1967), who studied an ecosystem in the Animas River, in Colorado. The accumulation of radium by insects, algae, and fish was directly proportional to radium concentrations (in water) varying between 0,1 and 2,2 pCi/L. Above this threshold (2,2 to 60 pCi/L), the accumulation potential of the organisms decreased.

1.2. Experimental Factors

Apart from the environmental factors described above, there are certain experimental factors that also affect the value of the coefficients. Most of the differences in the coefficients reported by the various authors could be explained by the fact that there are no experimental guidelines for the calculation of transfer coefficients. Authors should pay particular attention to the six following points:

(i) Fresh weight vs dry weight: Coefficients are obtained in reference to either fresh weight or dry weight. Blaylock (1982) suggests that fresh weight should always be used, since it makes it possible to estimate the rate of accumulation by live organisms in human food chains. Some authors have proposed the use of conversion factors for different types of weights. For instance, Sheppard and Thibault (1983), and Garten (1978), calculated that in 1 g of fresh vegetable tissue, there are between 0,20 and 0,25 g of dry weight. Sheppard and Thibault (1983) estimate that in 1 g of dry weight of vegetable tissue, there are about 0,02 g of ash; and that in 1 g of dry weight of soil, there are 0,96 g of ash.

(ii) Representative sample: The coefficients in the literature are often meaningless because the small number of measurements which they are based on does not allow the calculation of a significant mean. It would be preferable to determine the minimum number of measurements in order to arrive at statistically significant results.

(iii) Analysis and sampling errors: Errors such as those caused by unrepresentative samples and by experimental artifacts, contribute to the fluctuations observed in the coefficients derived by different authors.

(iv) Filtered and non-filtered solution: Some authors do not filter the water solution of aquatic samples (total concentration of the radionuclide) and others do (concentration of dissolved radionuclide). A filtered solution has a radionuclide concentration that is lower than that of the non-filtered solution because of a certain percentage of radionuclides that is bonded to the particles in suspension. Coefficients should be calculated on the basis of a filtered solution and a non-filtered solution.

(v) Distribution of the radionuclide in the tissues: Radionuclides are distributed differently in biological organisms (e.g., muscles, liver, bones, or fruits, roots and stems). Ideally, it would be preferable to determine the coefficients, not for a whole plant or animal, but for the various parts or organs consumed by man.

(vi) Chemical form of the radionuclide: The chemical form of the radionuclide is a significant factor in the accumulation of radionuclides by the organism. The chemical form of the radionuclide determines whether or not it forms bonds with the soil and sediment, as well as its capacity for ionic exchange with biological organisms. It is necessary to describe precisely the chemical form of the radionuclide that is being studied.

(vii) Taxonomic group: At present, the coefficients estimated by the various authors are classified in categories such as "Fish", "Invertebrates", etc. These categories are extremely vague. A better approach would be to classify the coefficients on the basis of more precise taxonomic groups (for example, "freshwater mollusks") and according to various environmental parameters.

Chapter II

BIOLOGICAL INTERPRETATION OF THE COEFFICIENTS REVIEWED

Table II and III show the range of coefficients of radionuclides from uranium and thorium decay chains in terrestrial and aquatic environments which were found in the literature. These tables can be useful for choosing transfer coefficients for environmental pathways analyses.

2.1 Aquatic Environment

Coefficients associated with the aquatic environment tend to vary for the same species from one location to another. This is due mainly to spatial and temporal fluctuations in environmental factors. This observation is characteristic of fresh waters, estuaries, and coastal waters of the ocean. The marine environment is relatively stable, therefore a greater uniformity in coefficients is expected. Finally, studies on aquatic species have been generally carried out *in situ*, so that the coefficients are generally realistic.

(a) Sediment

Several authors in Canada (Emerson and Hesslein, 1973; Hesslein and Slavicek, 1984), have shown that sediment accumulates significant quantities of radionuclides. The accumulation potential of the sediment depends upon the physico-chemical properties of the radionuclide under consideration and the characteristics of the aquatic environment. Thus, it is not surprising that there is a difference between the accumulation of radionuclides by sediments in fresh and salt waters. Canada has many oligotrophic lakes resulting from the typical geology. The water in these lakes has a low conductivity and is slightly alkaline. Under these conditions, sediments tend to accumulate significant amounts of radionuclides. Based on the coefficients proposed by IAEA (1982), the following sequence could be used to compare the accumulation potential of various radionuclides:

$$U = Ra < Pb = Po < Th$$

Table II - Transfer Coefficients Obtained from the Literature - Aquatic Environment

	Sediment FW	Plankton FW	Algae FW	Macrophytes FW	Insect Larvae FW	Invertebrates FW	Fish FW	Sediment SW	Plankton SW	Algae SW	Molluscs SW	Crustaceans SW	Fish SW
Th	n=2 22000 - 3000000	phyto n=2 8000 - 120000 zoo. n=1 3000	n=1 16210	n=1 232-8925	?	n=1 3000	n=4 (wf) 0,7-3281	n=1 1800-17000000	n=1 1250-20000	n=2 10-7700	n=1 30-300	n=1 30-300	n=1 100
	3000000					500	30			1000	1000	1000	10000
Po	n=1 100000	?	n=1 448	?	?	?	n=2 f=30-830 bone=138	n=1 120 - [illegible]	n=2 2500-970000	?	?	?	?
	10000					20000	[illegible]			1000	20000	20000	2000
Pb	n=1 180000 - 300000	?	n=1 198	?	n=1 49-16000	?	n=7 f=15-76 bone=150-1227 wf=4-1600	n=1 120 - [illegible]	n=1 24000	?	?	?	?
	10000					100	300			1000	100	100	100
Ra	n=7 10-730000	n=2 40-4420	n=3 40-22900	n=1 752-6900	n=1 50-9558	n=3 1-3900	n=13 f=3-27 bone=36-750 wf=11-1500	n=1 500	n=3 2750-40000	n=3 100-14000	n=1 6-5000	n=1 f=50 sh=630 wf=400-7000	n=4 wf=15-6000 f=45-950 eggs=120
	500					300	10			10	100	100	100
U	n=4 7,5-885000	phyto n=4 88-459 zoo.n=2 16-80	n=2 51-1576	n=1 62-215	n=1 0,6-522	n=3 2-33500	n=10 f=0,2-18 bone=20-189 wf= 0,4-36	n=1 0,6	n=2 50-170	n=2 10-700	n=1 400	n=1 400	n=1 20
	500					100	100			10	10	10	1

n = number of references
[x] = coefficient suggested by IAEA
wf = whole fish
f = flesh
sh = shell
? = no reference found in the literature
fw = fresh water
sw = salt water

Table III - Transfer Coefficients Obtained from the Literature - Terrestrial Environment

	Forage Plants	Food Crops	Wild Plants	Wine	Fm (cattle)	Fm (goats)	F (eggs)	Ff (cattle)	Ff (lamb)	Ff (pigs)	Ff (poultry)	Reindeer	Caribou	Elk	Moose	Deer	Beaver
Th	n=2 0,0008 - 0,002 0,003	n=1 0,005 0,0005	n=5 0,001-1	?	n=3 0,00005 - 0,0003 0.000005	?	?	n=1 0,0002 0,0001	?	?	?	?	n=1 f=0,0014	?	n=1 0,005	?	n=1 0,0003
Po	n=2 0,000002 - 0,2 0,004	n=2 0,0000012 - 0,0008 0,0002	n=1 0,03 - 0,2	?	n=6 0,00009 - 0,03 0.0001	n=2 0,18 - 0,45	?	n=4 0,01 - 0,05 0,003	n=1 0,04	?	?	n=3 f=0,05 liver=0,5 kidney=0,3 bone=0,4	n=2 f=0,05 - 0,23 liver=1,3 kidney=0,6 bone=0,9	n=1 f=0,004 liver=0,04 bone=0,03 kidney=0,02	n=1 0,3	n=1 f=0,003 liver=0,004 bone=0,03	n=1 0,015
Pb	n=4 0,0003 - 0,3 0,09	n=6 0,0001 - 0,02 0,01	n=2 0,08 - 2,8	?	n=4 0,000006 - 0,0001 0,0003	?	?	n=4 0,0002 - 0,001 0,0008	?	?	?	n=3 f=0,0005 liver=0,1 kidney=0,06 bone=0,6	n=1 f=0,008	n=1 f=0,0001 liver=0,004 kidney=0,006 bone=0,07	n=1 0,02	n=1 f=0,002 liver=0,003 bone=0,05	n=1 0,01
Ra	n=5 0,00006 - 0,6 0,2	n=12 0,0001 - 0,03 0,04	n=6 0,002 - 25	n=1 0,0008 - 0,001	n=10 0.0001 - 0.14 0,0006	?	n=1 yellow:0,3 white:0,1	n=4 0,0005 - 0,004 0,0005	n=1 0,003	n=1 0,001	?	n=2 f=0,003 - 0,03	n=3 f=0,001-0,05	?	n=1 0,02	n=1 f=0,008 liver=0,007 bone=0,03	n=1 0,0006
U	n=1 0,0001 - 0,1 0,01	n=3 0,00001 - 0,002 0,002	n=13 0,0003 - 8	?	n=5 0,00004 - 0,0006 0,0006	?	n=1 0,9-2,5	n=2 0,00007 - 0,0003 0,03	?	n=1 kidney=12 liver=0,03 f=0,06	n=1 1,2	?	n=1 f=0,002	?	n=1 0,03	n=1 f=0,2 liver=0,05 bone=0,09	n=1 0,0004

n = number of references
x = coefficient suggested by the IAEA
Fm = factor for milk in d/L
Ff = factor for meat in d/kg
? = no reference found in the literature
f = flesh

Thorium is heavily accumulated by sediment while uranium is comparatively less accumulated. The present review of coefficients in the literature shows the following sequence which was derived from the few studies found in the literature:

$$U = Ra < Pb = Po < Th$$

This sequence is similar to that obtained by IAEA. Thorium seems to have a distribution coefficient that is higher than the other radionuclides, although this observation has not been repeated by all authors (Brunskill et al. 1984).

(b) Aquatic Flora

Human beings do not consume many aquatic plants. It is nevertheless important to study them because they often form the base of food chains where man is the final consumer.

Aquatic plants, from the less evolved to the most evolved ones, concentrate significant amounts of radionuclides. Generally, plankton is the greatest accumulator. Phytoplankton accumulate radionuclides in an ionic form (whereas zooplankton accumulate radionuclides in the form of particles (Paschoa et al., 1982)). In these plants, and more highly evolved other groups as well, a large portion of the accumulation takes place by adsorption to the cellular walls.

Mahon (1982) found that plankton in non-contaminated environments accumulate a greater quantity of radionuclides than plankton in contaminated environments. No explanation for this observation was offered by the author. A possible explanation would be that the chemical form of the radionuclides in the contaminated environment is different from that of the radionuclides in the non-contaminated environment which is readily adsorbed to the cellular walls of the phytoplankton. Another explanation would be that there has been a disturbance in the metabolism of the plankton due to high concentrations of contaminants in the water, and that this has, in some way, prevented the accumulation of radionuclides.

The accumulation of radionuclides by plant species is different whether they be in fresh or saltwater environments. Pb-210 and Po-210 are much more concentrated in salt than in fresh waters (Table II). Polonium is actively selected by marine plants, and this observation is marked even more in the case of plankton, (Mahon, 1982)

The following sequences show the comparative accumulation of radionuclides by aquatic flora in fresh and salt waters:

Pb < Po < U < Ra < Th	(Freshwater environment)
U < Ra < Th < Pb < Po	(Saltwater environment)

(c) Aquatic Fauna

Two types of aquatic animals are examined: invertebrates and fish. The invertebrate group includes three main subgroups: insect larvae, marine crustaceans, and mollusks. Given that these three categories of invertebrates have very different metabolisms, their accumulation capacities differ. Therefore, it is necessary to obtain separate coefficients for each category.

Few studies have been done on insect larvae and marine crustaceans; those few show a wide range of coefficient possibly due to different modes of nutrition within these groups.

It is already well known that mollusks accumulate large amounts of heavy metals (Fretter and Graham, 1962). Most radionuclides of the uranium and thorium decay chains are heavy metals, and the coefficients obtained from the literature indicate that, in fact, mollusks accumulate large amounts of these radionuclides. In the light of some of the data collected (Table II), it seems that there are no qualitative differences between the accumulation by these animals in fresh and salt waters. Future studies should be undertaken to verify this observation. The behaviour of various radionuclides accumulated by mollusks can be summarized as follows:

fresh waters: comparison is not possible due to lack of data
salt waters: U < Th < Ra

Since no data were found on Pb-210 and Po-210, it was imprudent to reach any conclusions concerning these radionuclides. IAEA (1982) indicates that polonium is more concentrated than lead in the two environments.

Finally, judging by the number of studies found, fish are the most frequently studied species. Except for ethnic groups who grind the whole fish to make a type of pulp and for certain delicacies, man is mainly a consumer of fish flesh.

Radionuclides are dispersed through the various organs of the fish. Radium, uranium and lead have a tendency to concentrate in the bones, but this may vary with the physico-chemical conditions of the environment. In the female, a certain proportion of the radionuclides tends to accumulate in the eggs, so that, consequently, there is less concentration in the body of the fish itself. The distribution is less certain in the case of thorium and polonium.

Fish are greatly affected by environmental conditions, such as temperature, pH, salinity, conductivity of the water, etc. For example, Ophel (1978) found that in waters where conductivity is low fish accumulate a larger percentage of uranium and radium in their flesh. Swanson (1982) found that, in alkaline environments, radium is largely concentrated in the bones of the fish, but when the environment is acidic, this radionuclide tends to concentrate in larger quantities in the flesh. The salinity of the water also affect the coefficients obtained both in salt and fresh waters. The phenomenon of osmosis and the chemical form of the radionuclide in the two environments can also contribute to the differences observed between the coefficients. These differences indicate that fish from marine environments accumulate radionuclides to a greater extent than fish from fresh waters. In fact, Holzman (1967) has pointed out that Pb-210 is between three and five times more concentrated by saltwater species than by those living in fresh waters.

Qualitatively, the uptake of radionuclides in fish can be illustrated by the following sequence:

freshwater environment	U < Ra < Pb < Po
saltwater environment	U < Ra < Th

We see in Table II that transfer coefficients of thorium for fresh water fish vary widely (0,7 to 3 281). In marine environments, Polikarpov (1964) proposes a coefficient of 100, while IAEA (1982) suggests a coefficient of 10 000.

2.2 Terrestrial Environment

In the terrestrial environment, conditions are relatively stable; the coefficients obtained from the literature tend to vary less than those associated with aquatic environments.

There are many *in vitro* studies on terrestrial plant and animal species. Most wild animal studies have been conducted *in situ*. Even though laboratory studies make possible a better interpretation of the data, it is necessary to exercise some caution because they derive from conditions that may be more or less realistic. For example, injecting radionuclides to a cow can produce quite different results from those that are obtained from a situation in which the cow had consumed the same amount of radionuclides by grazing in a pasture.

(a) Terrestrial Flora

The present report will take into account only the transfer from soil to plant, since it has been assumed here that atmospheric deposits on the leaves are reduced to a minimum through the action of wind, rain, and habits relating to the preparation of food (Garten, 1978) (lichen is a special case where the atmospheric pathway is much more important than the soil pathway because of the small ramificated structure of this genus, the quasi inexistent root system and its long life.)

Lopatkina *et al.* (1970, *in* Sheppard and Thibault, 1983) found that only primitive plants accumulate radionuclides in a linear fashion. However, Sheppard and Thibault (1983) have demonstrated that this linearity also exists in the case of *Picea mariana* (Black Spruce).

Uranium is retained in the root system, and a small amount is transferred to the aerial portions of the plant, which may be eaten by animals. Thorium is transferred to the old sections of the aerial portions of the plant, such as the woody parts. The latter transfer is not of great consequence because very

few animals eat the woody parts of plants. Radium is transferred actively to plant leaves, and it is considered to be the most important radionuclide in terms of terrestrial transfer.

Wild plants seem to accumulate radionuclides the most (Table III). Osborn (1965) found that this phenomenon is even more pronounced for the accumulation of uranium by bog plants, thorium being accumulated to a lesser extent. Forage plants and food crops accumulate lesser amounts of radionuclides and the coefficients associated with food crops are characterized by their stability from one environment to the next.

The comparative uptake of radionuclides based on the coefficients obtained from the literature, can be defined in terms of the following sequence:

forage plants	Th < Pb = Po < Ra
food crops	Po < U < Pb < Ra
wild plants	U < Pb = Po = Th < Ra

(b) Terrestrial Fauna

In this section, coefficients associated with domestic animals will be discussed separately from those associated with wild animals.

The domestic animal that is studied the most is the cow. Studies on the transfer of radionuclides to cow milk are relatively numerous. There are fewer studies on the transfer of radionuclides to beef. The comparative uptake of the various radionuclides has been summarized on the basis of data collected on milk and beef.

Fm (transfer coefficient to cow's milk)	Pb = Th < Po = U < Ra
Ff (transfer coefficient to beef)	U < Ra = Th = Pb < Po

Thus, radium is the radionuclide most readily transferred to milk, while polonium seem the one most readily transferred to beef.

Table II shows that other farm animals accumulate radionuclides more than cows. For example, polonium is transferred in larger amounts to goat milk (Johnson and Watters, 1972). It seems that rumination is more efficient in the goat, and that this makes possible a larger accumulation of polonium. The transfer of polonium and radium to lamb's meat (McDowell-Boyer et al., 1979) is of the same order as that to beef. The transfer of radium to pork meat is similar to the transfer to beef, but the transfer of uranium to pork meat is much higher than the transfer to beef. McDowell-Boyer et al. (1979) found that 70% of the amount of radium ingested by pigs is accumulated in the muscles, while 97% of the amount of radium ingested by lambs is accumulated in the bones. Finally, the transfer of uranium to chicken meat is clearly higher than the transfer to beef (Prister, in Garner, 1972).

In Russia, a study on the importance of uranium contamination of various food products (Prister, in Garner, 1972) produced the following sequence:

bread > cereals > dairy products > meat > potatoes and vegetables > eggs > milk.

In North America and Europe, the consumption of wild animals by man is generally much lower than that of domestic animals. Nevertheless, the contamination of wild animals can become an important factor for certain populations. Animals such as caribou, moose, deer, beaver, wildfowl and small game, are hunted for sport and for livelihood.

In general, wild game is more fatty and less meaty than that of domestic animals. The transfer of radionuclides seems less efficient in these animals than in domestic animals, and this is confirmed by the data in the literature. However, two exceptions were noted: the transfer of uranium to deer and moose meat is higher than the transfer of uranium to beef. The reason behind this is obscure.

The accumulation of radionuclides by wild animals is compared in the following sequences: (The reader should be reminded that most sequences are derived from only a few results and therefore should be interpreted cautiously.)

Th: caribou = moose > beaver

Po: moose > beaver > reindeer = caribou > elk = deer

Pb: moose > beaver > deer > elk = caribou

Ra: reindeer > moose > caribou = deer > beaver

U: deer > moose > caribou > beaver

2.3 Comparative Uptake of Radionuclides

In Tables II and III, some of the coefficients suggested by IAEA (1982) are significantly lower than those found in the present literature search. These coefficients pertain to the following categories:

Sediments:	Ra
Freshwater invertebrates:	U
Freshwater fish:	Pb
Food crops:	Th
Fm (cow):	Th, Ra
Ff (beef):	Po

The description of the comparative uptake of radionuclide formulated by IAEA and this report, as shown in Table IV is very similar, say for the one exception: IAEA indicates that uranium is transferred more readily to beef than the other radionuclides whereas the present report indicates that uranium is the least transferrable (polonium was the radionuclide most readily transferred). The cause of the disparity between both references is unclear.

An examination of Table IV could lead to the following generalities:

(a) Uranium and radium are not readily absorbed by sediment, while thorium is more absorbed.

(b) Uranium and radium are very mobile in terrestrial transfer and are not heavily accumulated in aquatic environments.

Table IV - Comparative Uptake of Radionuclides

(R) on the basis of this report (I) on the basis of IAEA (1982) data

Category	Comparative Sequences	Comments
Sediment:	(R) U = Ra < Pb < Po < Th (I) Ra = U < Pb = Po < Th	fresh water
Aquatic flora:	(R) Pb < Po < U < Ra < Th (I) * * * * *	fresh water
	(R) U < Ra < Th < Pb < Po (I) U < Ra < Pb = Po = Th	salt water
Invertebrates:	(R) no comparison possible (I) U = Pb < Ra < Th << Po	fresh water
	(R) U < Th < Ra (I) U < Ra = Pb < Th << Po	salt water
Fish:	(R) U < Ra < Pb < Po < (I) U < Th < Ra = Po < Pb	fresh water
	(R) U < Ra < Th (I) U < Ra = Pb < Po << Th	salt water
Forage plants:	(R) Th < Pb = Po < U (I) Po = Th < Pb = U < Ra	
Food crops:	(R) Po < U < Th < Pb = Ra (I) Po = Th < U < Pb = Ra	
Wild plants:	(R) U < Pb = Th = Po < Ra (I) * * * *	
Fm (cow):	(R) Pb = Th < Po = U < Ra (I) Th < Po = Pb = Ra = U	
Ff (beef):	(R) U < Ra = Th = Pb < Po (I) Pb = Ra = Th < Po < U	disparity between (I) and (R)

(c) The opposite is true of thorium and polonium. They are not very mobile in terrestrial environments but are very mobile in aquatic environments.

(d) Lead usually shows intermediate values, except for freshwater fish, which concentrate large amounts of this radionuclide.

(e) The radionuclides are more readily adsorbed to the sediments in freshwater environments than in the saltwater environments.

(f) Freshwater plants accumulate radionuclides more readily than marine species, except for polonium, where the opposite is true.

(g) Invertebrates in the two environments accumulate radionuclides in the same way, except for uranium, which is more concentrated in freshwater environments.

(h) Fish accumulate radionuclides more readily in marine environments, except for lead, which seems to be more concentrated in fresh water environments.

Finally, the ranges of coefficients proposed in this report, as well as the general hypotheses formulated regarding the uptake of radionuclides, are only an attempt at generalization. Given the scarcity of data, many statements made in this report remain to be tested and proven. Nevertheless, the information given should be useful in assessing radionuclide transfers when undertaking environmental pathways analyses.

CONCLUSION

There are many uncertainties and varying factors associated with transfer coefficients which were outlined in Part I of the present report; the use of these coefficients should, therefore, be prudent and only made of when site-specific information cannot be reasonably obtained.

This report has presented a table which gives the ranges of transfer coefficients representing the accumulation potential of radionuclides in aquatic and terrestrial organisms. General statements have also been formulated on the transfer characteristics of the radionuclides. Together, this information can be used to select appropriate coefficients for pathways analysis models that would take into account the specific aspects of the site under consideration.

Finally, readers should be aware that more studies in this area are being conducted abroad and in Canada. Pathways analyses are now seen as "fashionable" and new data are being published. The user of transfer coefficients, in addition to finding useful information in this report, should also refer to the most recent literature.

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APPENDIX A - Physico-Chemical Characteristics of Radionuclides from U and Th Decay Chains

Radionuclide Characteristic	U-238	Th-232	Ra-226	Po-210	Pb-210
Physical half-life (ICRP-38)	$4,5 \times 10^9$ a	$1,4 \times 10^{10}$ a	1620 a	138 d	22,2
Biological half-life and target organ (ICRP-30)	bone = 20d kidney = 6d other tissues = 6d	bone = 8000d liver = 700d other tissues = 700d	bone = 10-12 a other tissues = 900d	liver = 50d spleen = 50d kidney = 50d other tissues = 50d	bone = 10000d liver = 10000d kidney = 10000d other tissues = 1000d
Type of radiation, energy (ICRP-38)	α 4,20 MeV (75,3%) α 4,15 MeV (25%)	α 4,01 MeV (77%) α 3,95 MeV (23%)	α 4,78 MeV (94,3%) α 4,59 MeV (5,7%) α 0,19 MeV (4%) α 0,26 MeV (0,01%)	α 5,30 (100%)	β 0,015 MeV (81%) β 0,06 MeV (19%) β 0,05 MeV (3,9%)
Chemical characteristics (CRC Handbook of chemistry and physics.)	Forms generally related to the nuclear industry are insoluble.	Thorium oxides and hydroxides are not soluble. Nitrates, sulfates, chlorides and perchlorides are soluble.	Generally soluble.	Tendency towards the formation of radiocolloids. Chemical form will greatly influence its transfer through the environment.	Behaviour similar to rare earths.

APPENDIX B Biological and Ecological Characteristics of Radionuclides from U and Th Decay Chains

Radionuclide Characteristic	U-238	Th-232	Ra-226	Po-210	Pb-210
Biological characteristics (ICRP-30)	Since the specific activity of uranium is low, the chemical lesions to the kidneys are more hazardous than the radiological effects.	Tendency to concentrate in the surface of the bones.	It is deposited on the bones but not uniformly. Following the decay of Ra-226 in the bones, 67% of the resulting Rn-222 is diffused in the blood stream and exhaled.	Uniform distribution in the body.	Competes with calcium at absorption sites in bones.
Ecological characteristics (This report)	Uranium is absorbed in relatively small amounts by aquatic organisms, whereas it is readily absorbed by plants at the level of the roots.	Very mobile in aquatic environments, not mobile in terrestrial environments because it forms strong bonds with the soil.	Radium is easily absorbed by plants. However, organic matter in soil binds to Ra which will be less easily transferred to the plant. Generally, Ra is easily transferred to aerial parts of the plants.	It is especially significant through the atmosphere pathway to certain plants (e.g. lichen, tobacco).	Same as Po-210.

GLOSSARY

APPENDIX C

Acute exposure:	Exposure during a short time period, generally less than 96 hours (contrary of acute = chronic)
Abiotic and biotic factors:	Environmental factors that can have a direct effect on living organisms. Abiotic factors include: climate, soil characteristics, the physico-chemical composition of water. Biotic factors include factors such as predatory behaviour, competition and parasitism, that are endogenous to the species (nutritional characteristic, growth, etc.).
Homeothermic:	An animal whose body temperature is constant (birds and mammals).
Lichen:	Plants that live on the soil, trees and stones, consisting of a flat or branching thallus, or living in association with an alga and a fungus.
Trophic level:	Organisms are said to belong to the same trophic level when they are separated by chlorophyllic plants by the same number of stages in the food chain. By definition, chlorophyllic plants represent the first trophic level.
Oligotrophic lakes:	Deep lakes that contain small amounts of nutrients; they are very clear, and contain large amounts of dissolved oxygen.

Plankton: Group of microscopic plants (phytoplankton) or animals (zooplankton) living close to the surface of the water, and more or less drifting with the currents.

Poikilothermic: Animals whose internal temperature varies with that of the environment, such as reptiles and fish.

Peat bog: Acid swamp containing sphagnum, hypnea, drosera, etc.

Rumination: Particular digestion made by ruminants who swallow the non-chewed hay then bring it back into the mouth where it is masticated then sent it back down for a gastric digestion.